Impact of Neutron Irradiation on the Thermoelectric Properties of Rare Earth-based Thermoelectric Materials

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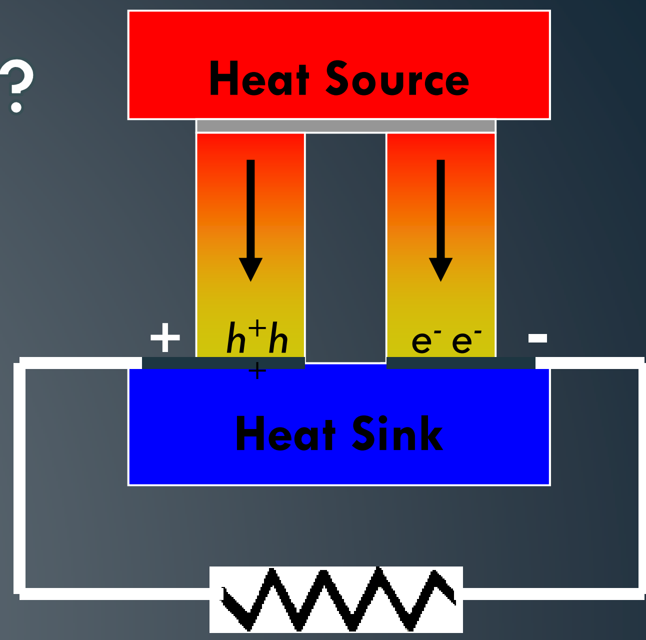
Outline

- What are thermoelectrics?
- NASA's uses
- Life time performance criteria
- The irradiation
- Research method and data
  - Resistivity
  - Thermal Conductivity
- Conclusion

What are Thermoelectrics(TE)?

- Conversion of heat into electric energy.
- Done based on the Seebeck Effect.
- Temperature differential causes electrons to diffuse to the cold side. (Potential barrier that arises to prevent diffusion is the Seebeck voltage.)
- The connection of p- and n-type elements creates a power device.



Carnot | TE Materials

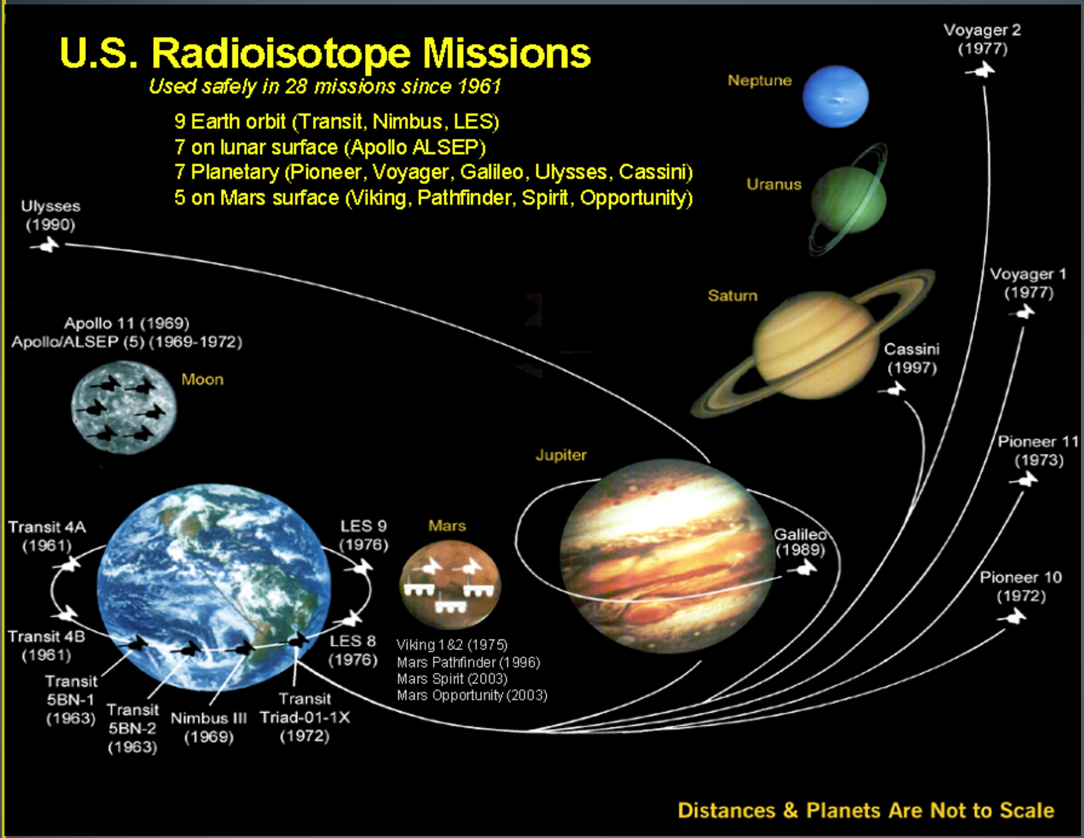
$$\eta_{\max} = \frac{T_{hot} - T_{cold}}{T_{hot}} \cdot \frac{\sqrt{1+ZT} - 1}{\sqrt{1+ZT} + \frac{T_{cold}}{T_{hot}}}$$

$$zT = \frac{\alpha^2 T}{\rho k}$$

α = Seebeck coefficient
T = Temperature
ρ = resistivity
k= Thermal conductivity
All of which are testable in the lab

Conversion efficiency is function of ZT and ΔT

Why use thermoelectrics to power spacecrafts?



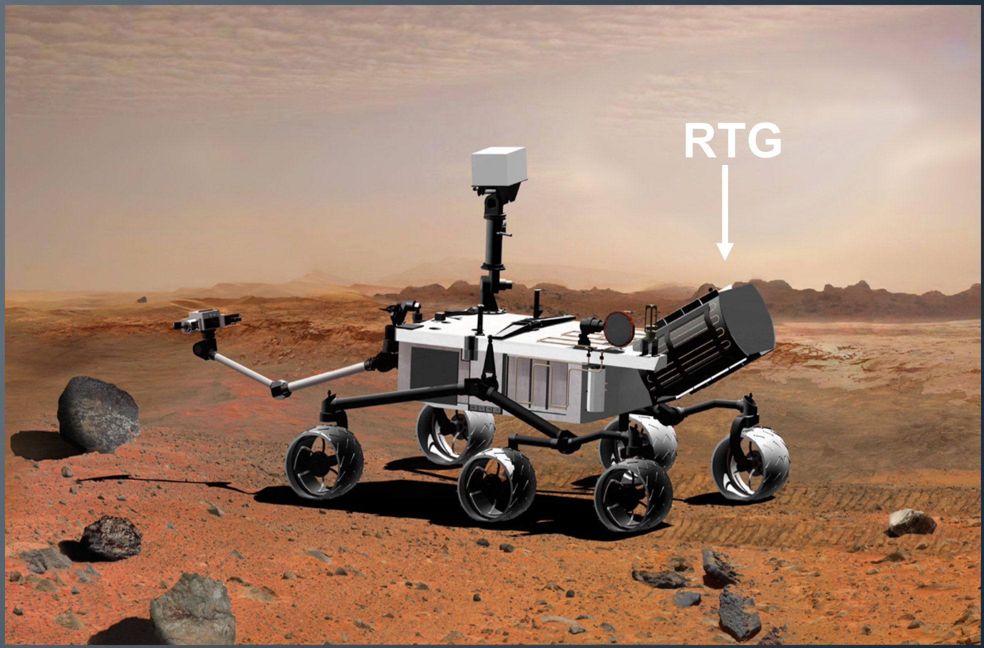
- NASA's deep space and planetary missions
  - Where solar power is not available or not practical.
- Compact, solid-state devices
  - Survives the vibrations from launch.
  - No vibration or electromagnetic interference for sensitive instrumentation.
- Long lifetimes
  - Voyager over 30 years

Current NASA Missions

- The thermoelectric power systems used are called Radioisotope Thermoelectric Generators (RTGs) which power deep space probes and rovers.

Cassini - Saturn

http://saturn.jpl.nasa.gov/;



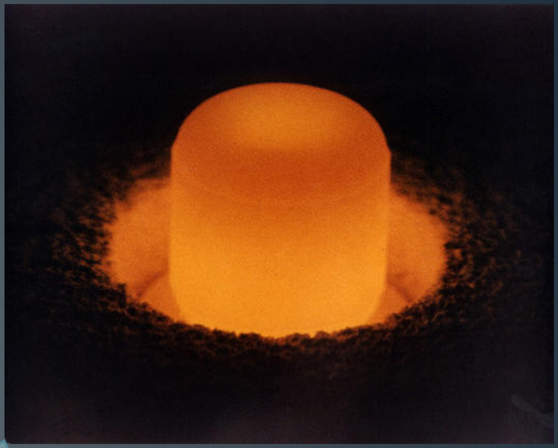
Mars Science Laboratory

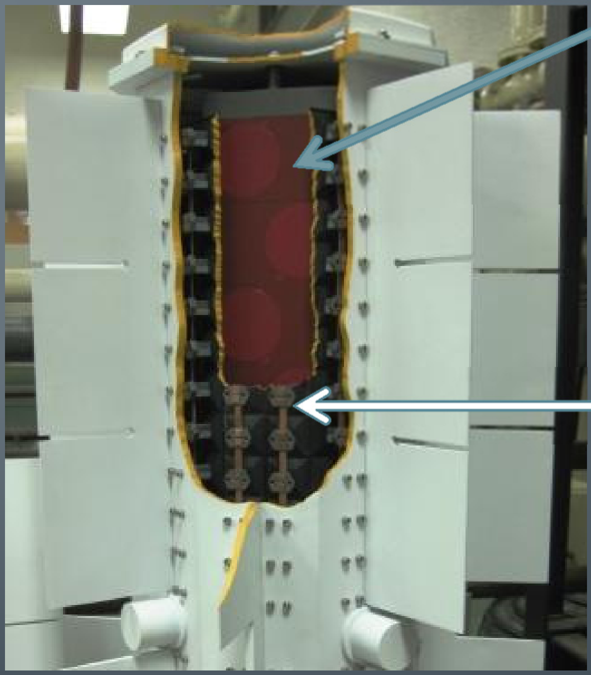
http://marsprogram.jpl.nasa.gov/msl/

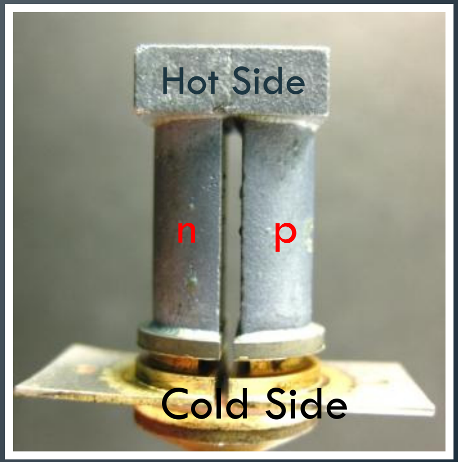
Hot Side?

- The hot side of RTG's is typically kept at temperatures around 1275K. How???
- Through alpha decay based heat sources.
- Pu-238 is the heat source used.

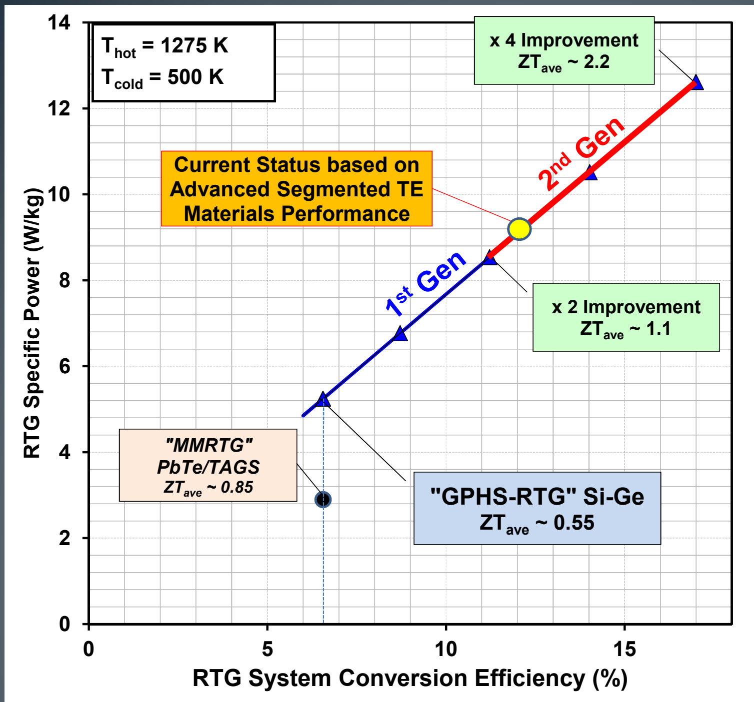
Hot Pu-238

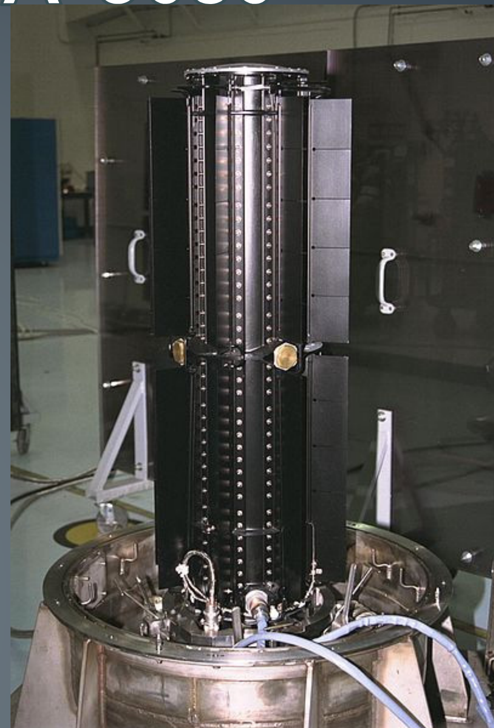


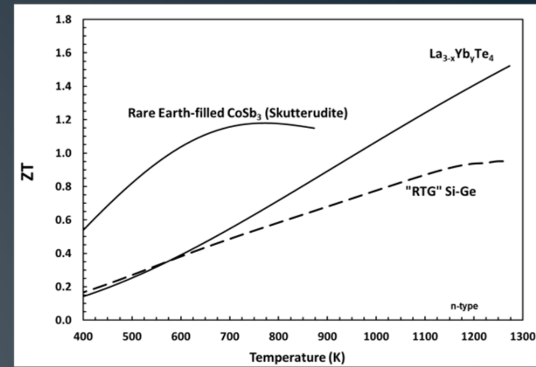


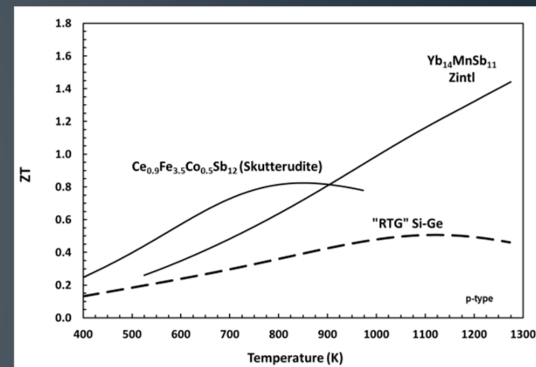


Cont. Current NASA uses

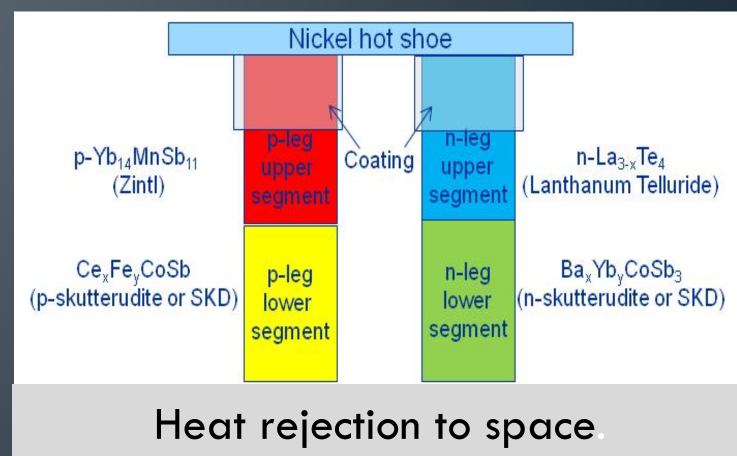








- Under ATEC project, high-temperature TE materials are under development for integration into Radioisotope Thermoelectric Generators (RTGs) at the Jet Propulsion Laboratory (JPL).



1) http://mediaarchive.ksc.nasa.gov/detail.cfm?mediaid=521
2) http://marsprogram.jpl.nasa.gov/msl/multimedia/images/?ImageID=3504

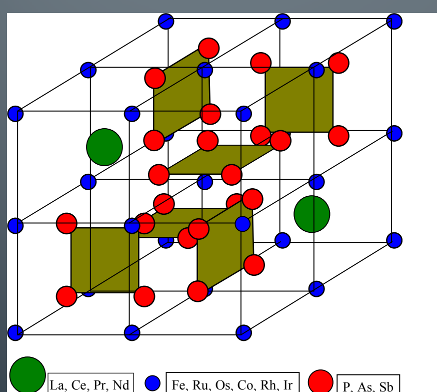
Demonstrating Life-time of New TE Technology for use in RTGs.

- Determine potential problems which could lead to the degradation of RTG's power supply over time:
  - Sublimation of materials
  - Breakdown of interfacial electrical contact resistance
  - Degradation of thermoelectric properties
  - Mechanical behavior

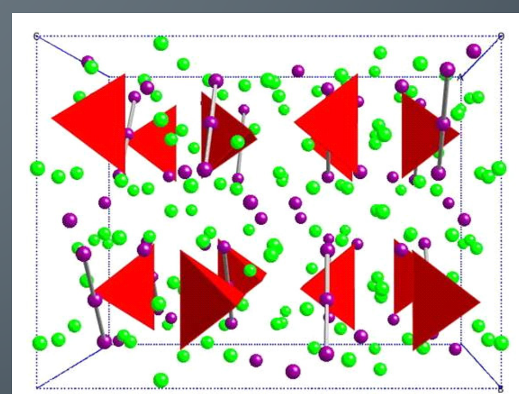
Potential impact of irradiation on thermoelectric materials!

Possible Performance Degradation through Irradiation

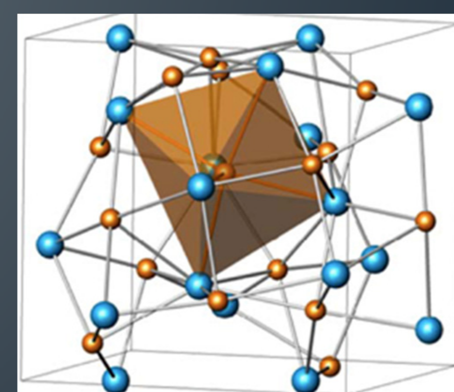
- Undesired contaminating Pu-240 in Pu-238 may cause a degree of fission neutron radiation over time.
- Fission neutrons in the energy range of 100KeV to 8MeV could cause lattice damage in TE materials.
  - Some of this damage may be annealed in real time during operation.
- Any change in the lattice structure may effect the TE properties of the materials used.
  - This would decrease the performance of RTG’s over time.



Skutterudites Lattice



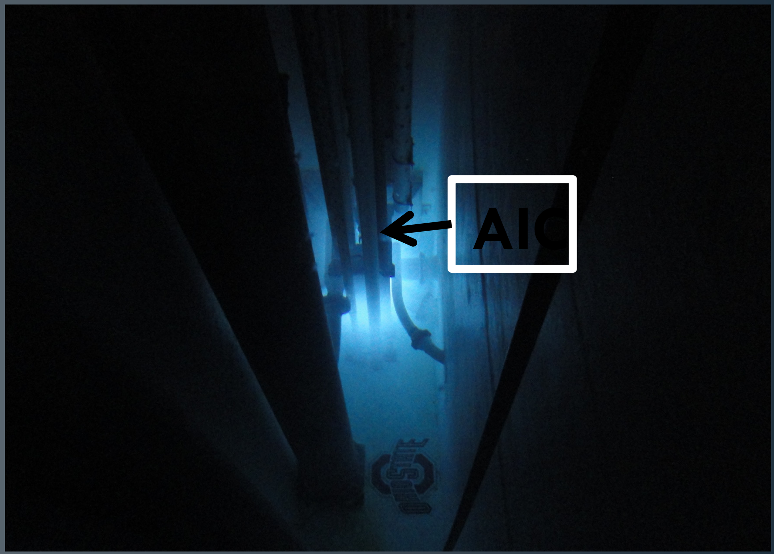
Zintl Phases Lattice



$La_{3-x}Te_4$

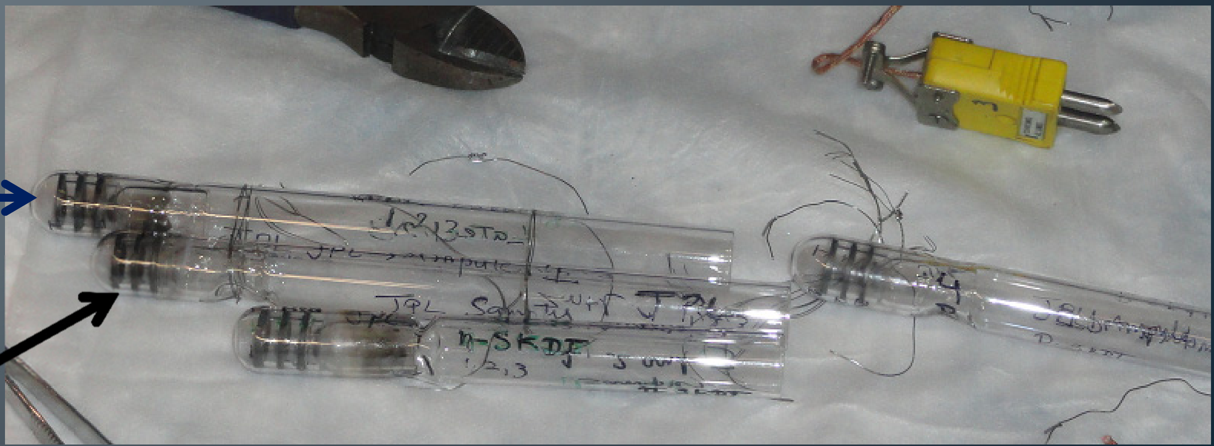
Simulated Irradiation

- The below samples were exposed to 17 years worth of neutron radiation in 35 minutes. This was done near room temperature at the Ohio State University Research Reactor (OSURR).
  - 3 Samples: n-type $La_{3-x}Te_4$
  - 3 Samples: p-type $Yb_{14}MnSb_{11}$ (Zintl)
  - 3 Samples: n-type filled skutterudites
  - 3 Samples: p-type filled skutterudites



3 samples
per ampoule

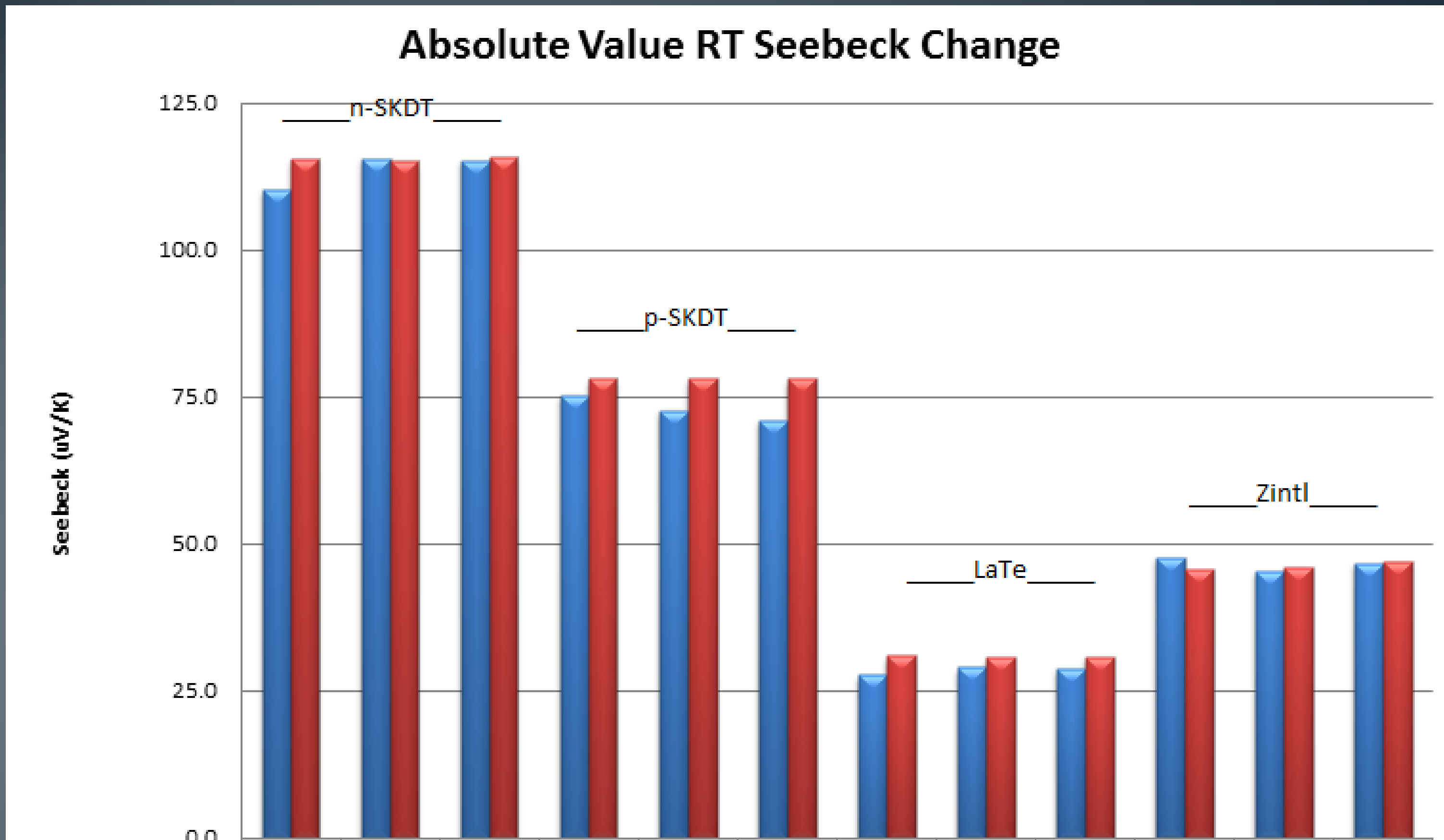
Samples separated
Inside the ampoule
By quartz separators



Measurements

- The TE properties of the materials were tested at two different stages
  - Pre-irradiation at room temperature (RT)
  - Post-irradiation RT and high temperature (HT) measurements.
- The properties tested were:
  - Electrical resistivity (ρ)
  - Thermal conductivity (K)
  - Seebeck coefficient (α) (pending)

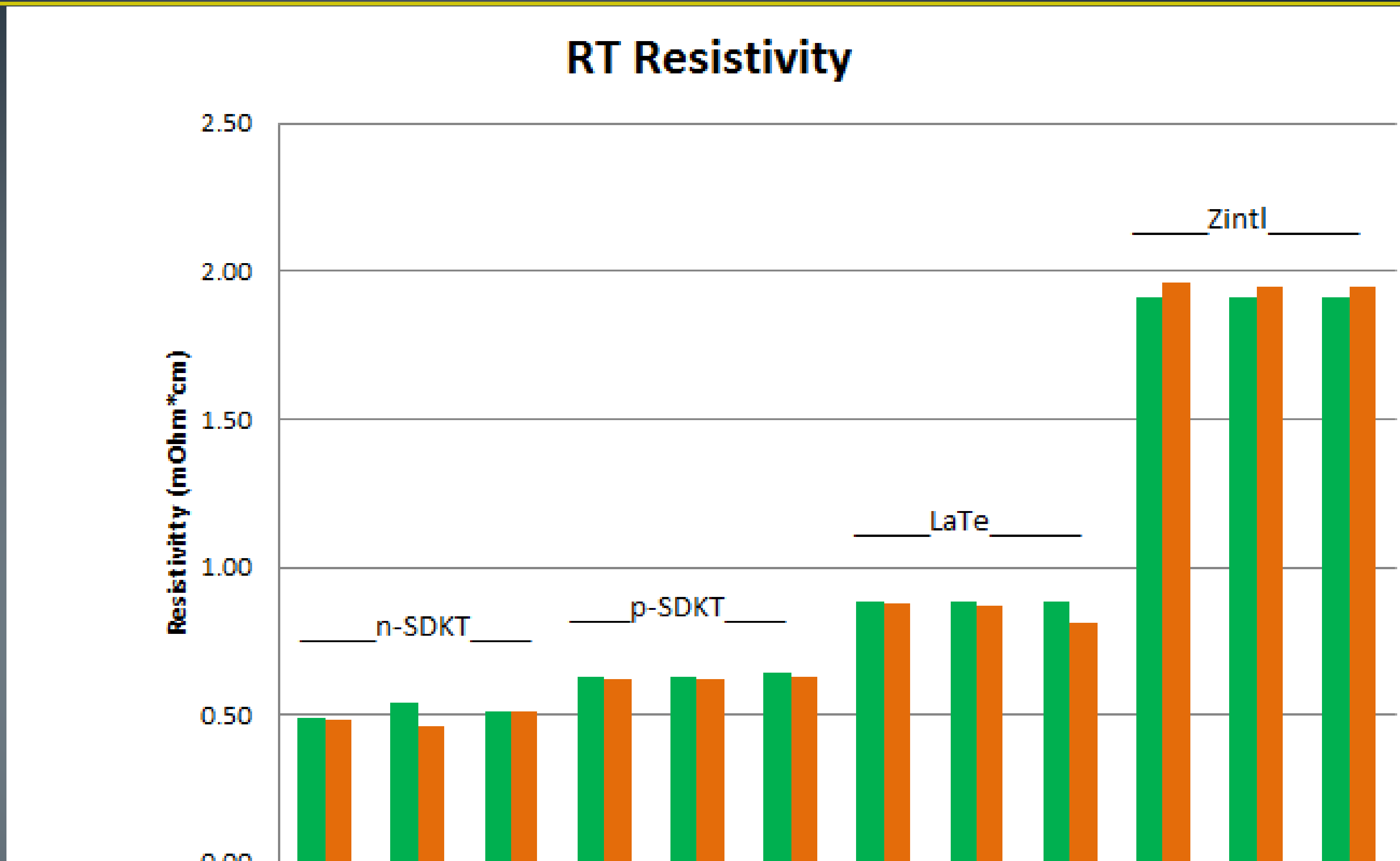
$$zT = \frac{\alpha^2 T}{\rho k}$$



	1	2	3	4	5	6	7	8	9	10	11	12
Post-Irradiation	110.4	115.6	115.2	75.2	72.8	71.1	27.8	29.1	29.0	47.7	45.5	46.8
Pre-Irradiation	115.6	115.0	115.7	78.0	78.0	78.0	31.0	30.6	30.8	45.7	46.1	47.0

Seebeck change was all under 10%

- Within margin of error for RT Seebeck



	1	2	3	4	5	6	7	8	9	10	11	12
Pre-Irradiation	0.49	0.54	0.52	0.63	0.63	0.64	0.88	0.88	0.88	1.91	1.91	1.91
Post-Irradiation	0.48	0.46	0.51	0.62	0.62	0.63	0.88	0.87	0.81	1.96	1.95	1.95

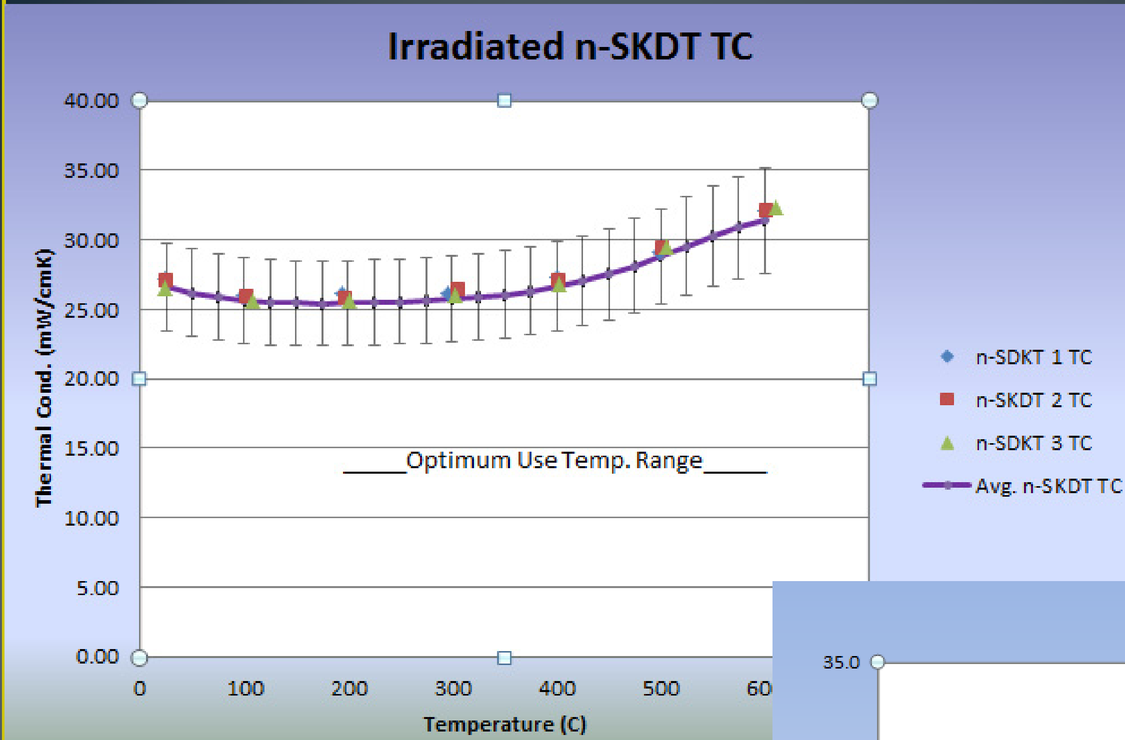
Resistivity change was within 5%

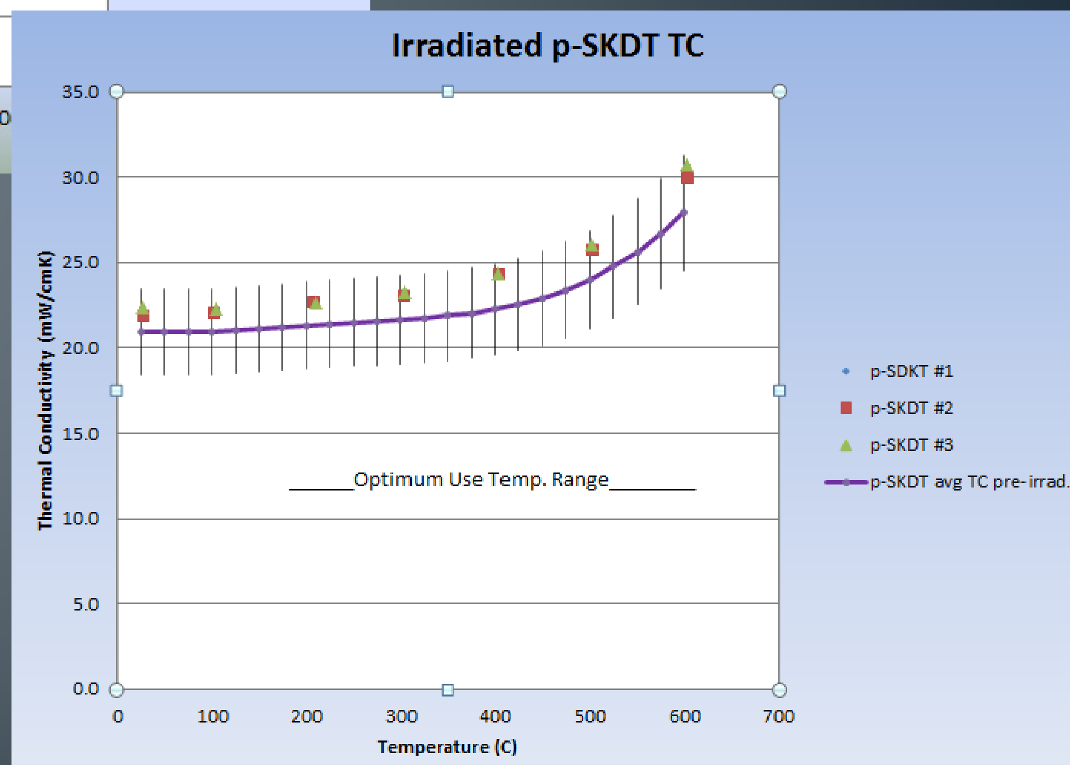
Within margin of error for RT Hall measurements

RT Electrical resistivity did not degrade!

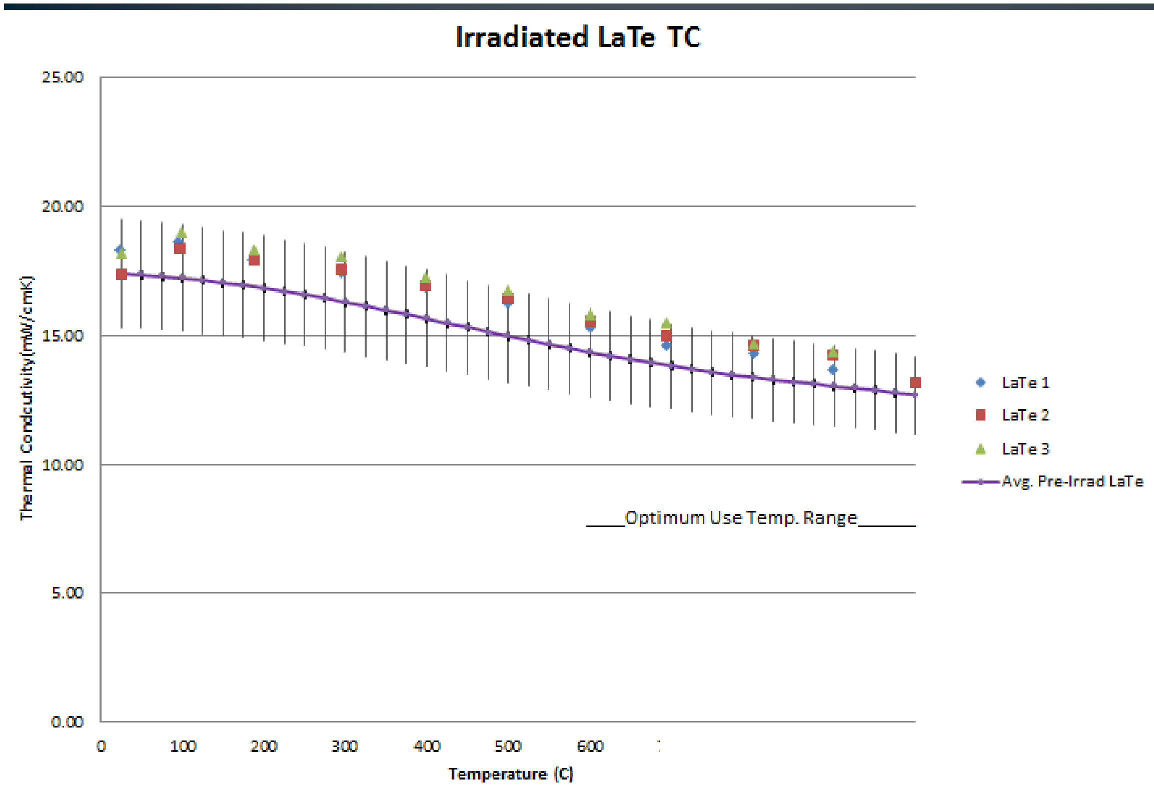
Thermal Conductivity (TC) Measurements

Post-Irradiation Thermal Conductivity was compared to pre-irradiation average
- Avg. has 12% spread.



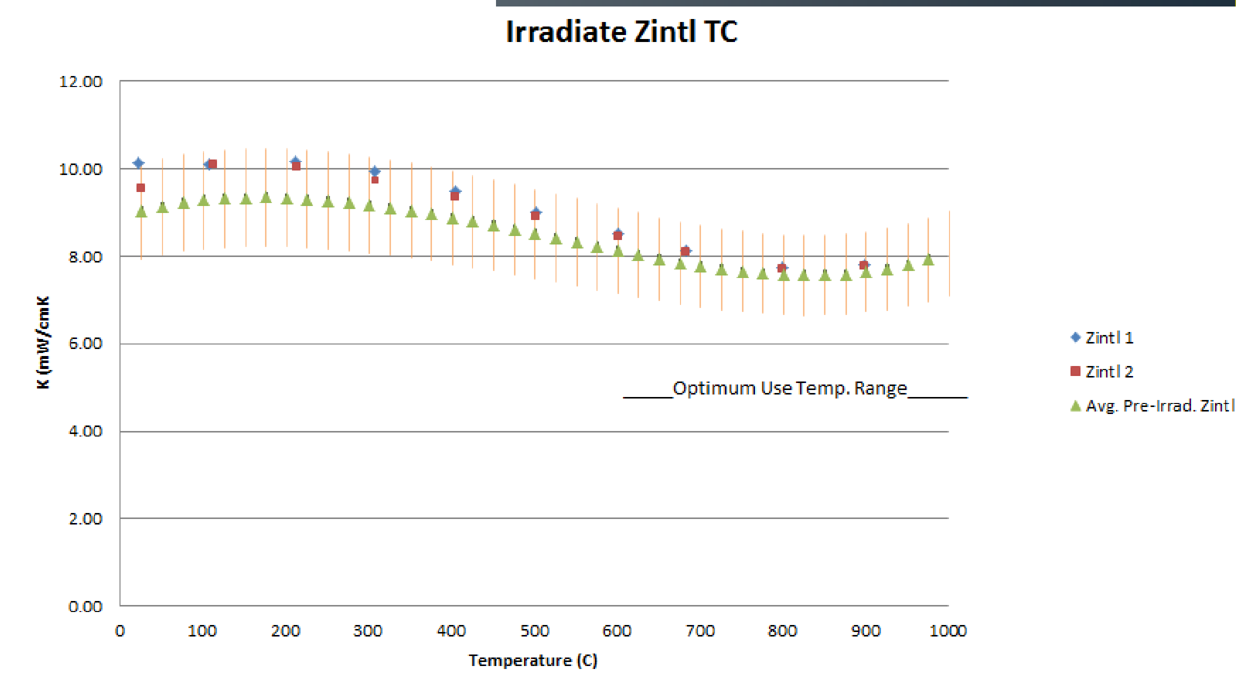


All TC data measured post irradiation was within the spread of the pre-irradiation data, as well as within measurement error.

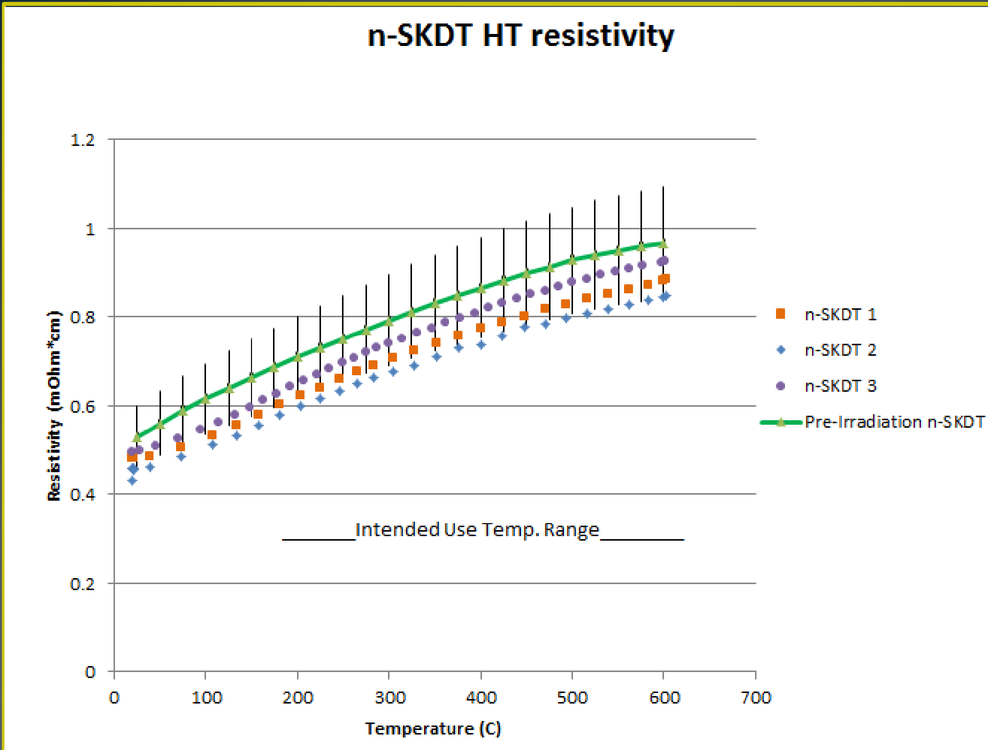


Post-Irradiation Thermal Conductivity was compared to pre-irradiation average

- **Avg. has 12% spread.**



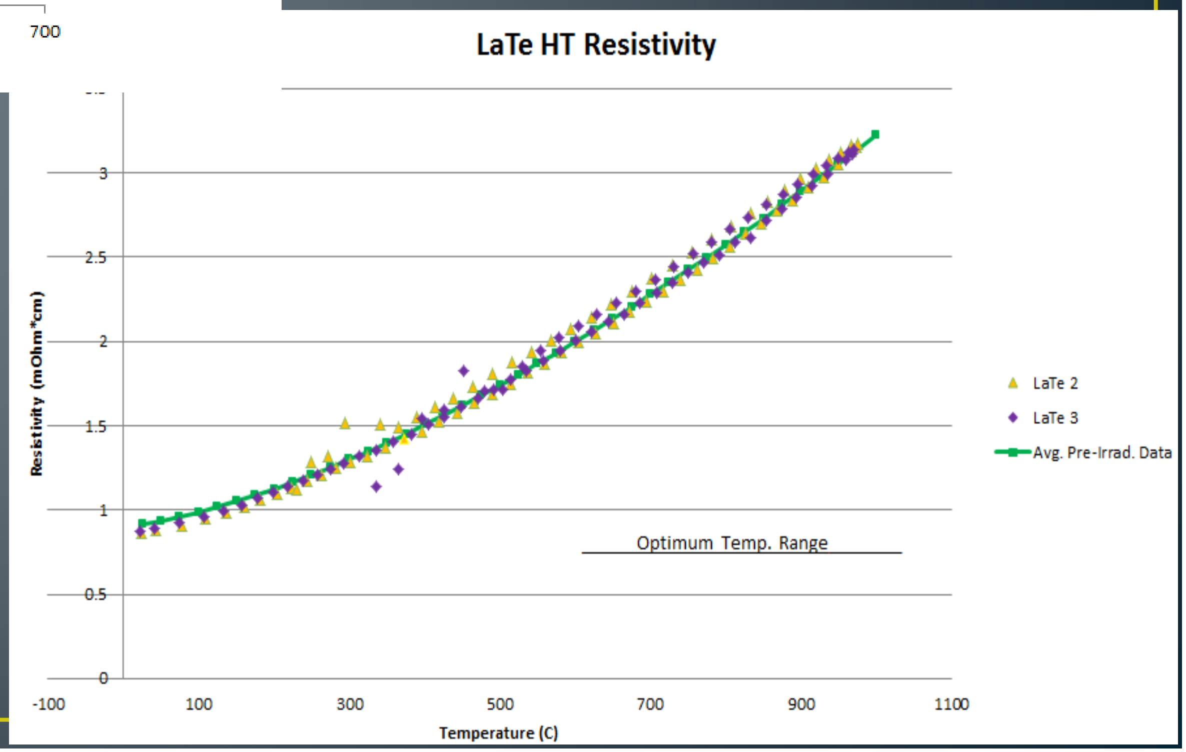
TC data points fell within the spread of the pre-irradiation data points.

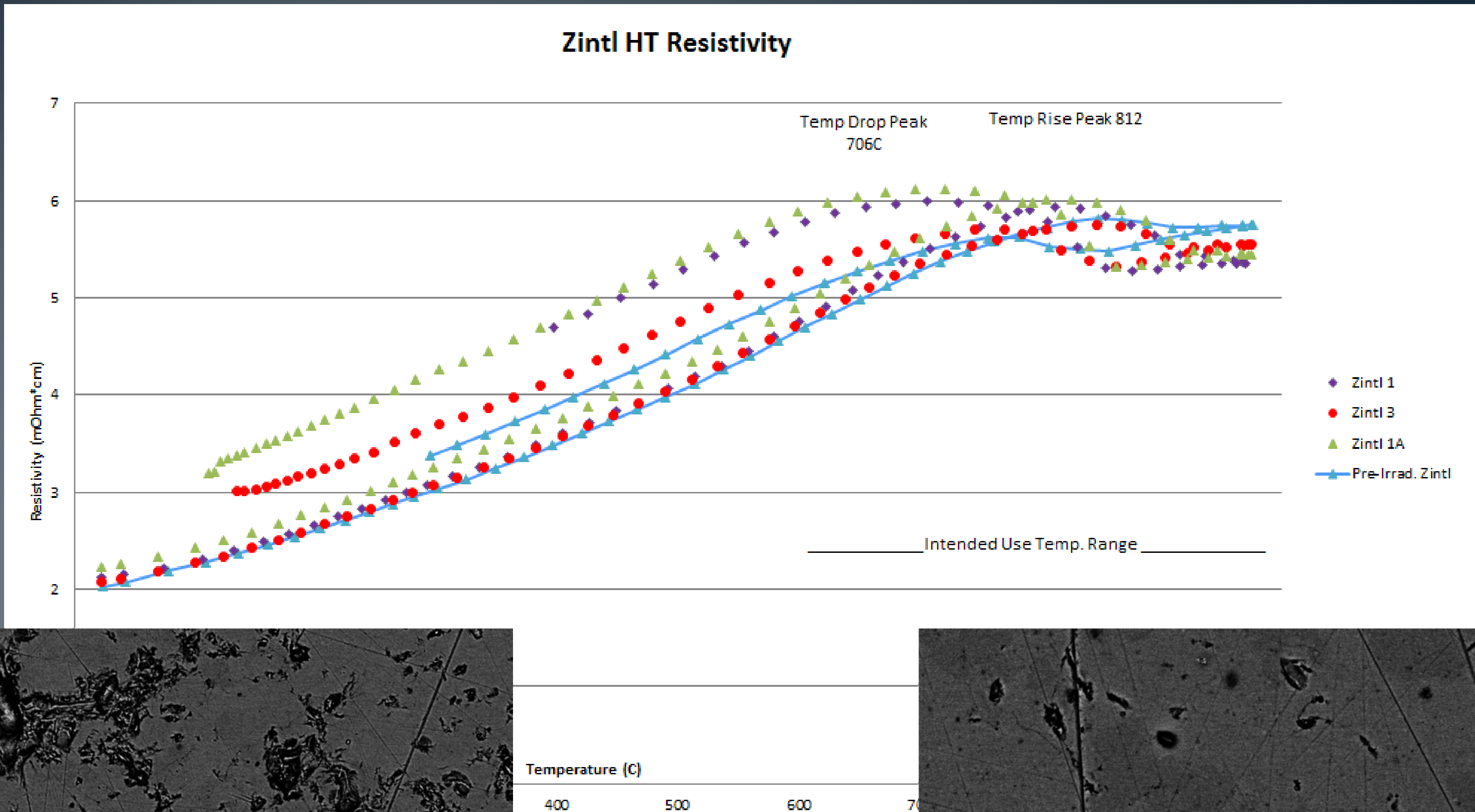


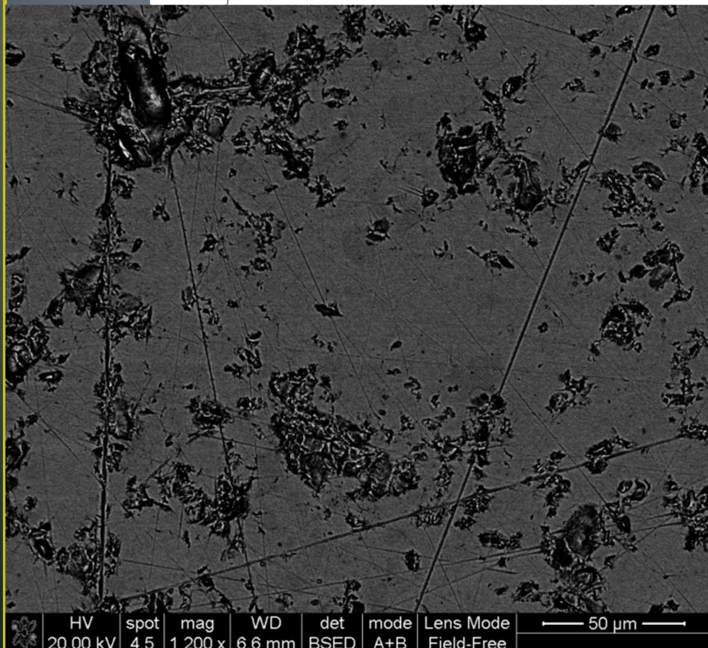
LaTe followed the Pre-Irradiation curve almost exactly.

- No HT degradation.

- Sample 1 and 3 fell within the spread of the pre-irradiation data
- Sample 2 was just beyond it. Accounting for error on each individual measurement Sample 2 may still fall within spread.

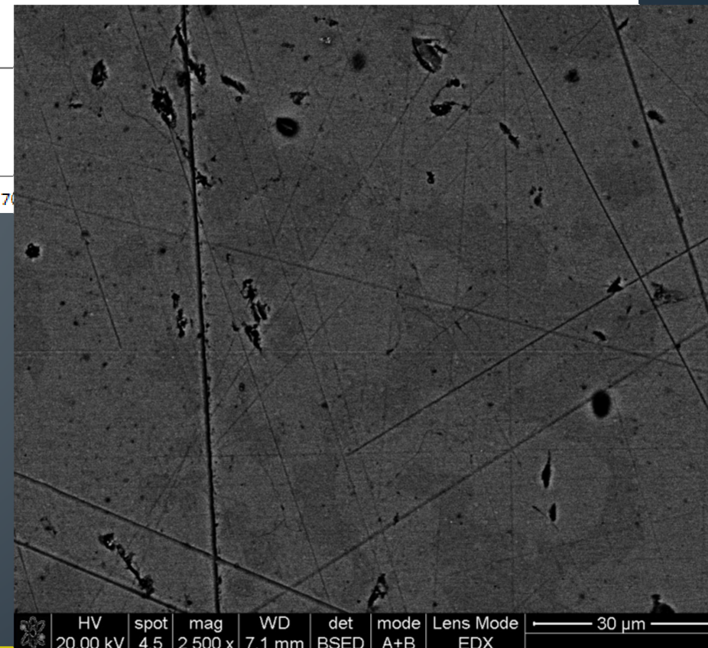






Left: Non-irradiated Zintl

Right: Irradiated Zintl



Analysis

- n-Skutterudite, p-Skutterudite, and LaTe post-irradiation resistivity and thermal conductivity all were within the margin of error or spread of pre-irradiation measurements.
- Due to contamination in the original Zintl sample batch, the behavior of post-irradiated data was skewed.

Conclusion

- No explicit radiation damage could be measured in TE properties of n- and p-Skutterudites.
  - Pending Seebeck measurements
- LaTe: Displayed no noticeable change in TE properties.
- Zintl: May have been influenced from the neutron irradiation, but original sample batch was contaminated, thus new samples need to be created and tested.
- Overall it seems these materials will be able to resist degradation to any neutron radiation.

- <u>Further research </u>:
  - HT Seebeck should still be recorded for all of the samples.
  - Zintl had contaminates in the original sample and needs to be remade.

Acknowledgments

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The Advanced Materials Group

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